Inhomogeneous ground state and the coexistence of two length scales near phase transitions in real solids

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Real crystals almost unavoidably contain a finite density of dislocations. We show that this generic type of long–range correlated disorder leads to a breakdown of the conventional scenario of critical behavior and standard renormalization group techniques based on the existence of a simple, homogeneous ground state. This breakdown is due to the appearance of an inhomogeneous ground state that changes the character of the phase transition to that of a percolative phenomenon. This scenario leads to a natural explanation for the appearance of two length scales in recent high resolution small-angle scattering experiments near magnetic and structural phase transitions.

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The modern theory of phase transitions (PTs) in pure and weakly disordered crystals is based on the scaling hypothesis and the renormalization group (RG). It has been confirmed by a large number of experiments and numerical simulations.

Therefore, the results of some recent high resolution X–ray and neutron scattering experiments for various crystals near structural and magnetic PTs were quite unexpected [1]: in contrast to the predictions of conventional scaling theory two distinct large scales were observed. The temperature dependence of the smaller scale was found to be broadly consistent with the results of the conventional theory for the correlation scale of thermal fluctuations. Quite recently explicit experimental evidence for Verneuil grown samples of $SrTiO_3$ [2] confirmed earlier speculations that the second, larger scale is connected with the presence of defects in a very large region near the surface.

All experiments that found two length scales used crystals which were considered to be of good quality. Nevertheless, surface preparation creates a significant density of defects in a surface layer with a thickness up to several μ [1–4]. Based on this observation, Altarelli et al. suggested [5] that most of the defects are actually dislocation dipoles which induce elastic strains with a pair correlator $G(\mathbf{r}) \sim |\mathbf{r}|^{-2}$. They tried to explain the critical behavior (CB) in the disordered layer as that of the Weinrib–Halperin (WH) model [6]. The coexistence of two scales was then interpreted as a simple superposition effect, with scattering from the larger scale in the disordered layer (where the CB is governed by the WH fixed point) and from the smaller scale (corresponding to conventional CB) in the bulk of the sample. However, later experiments using very thin Holmium films found clear evidence that near the magnetic PT temperature both scales coexist in the same volume fraction of the sample [7].

By construction, the conventional RG procedure is not able to explain the *simultaneous* existence of two length scales. Not only its detailed value and temperature dependence, but even the principal theoretical basis for the origin of the second length scale appears to be unclear.

In this Letter we point out the shortcomings of the RG procedure for the treatment of systems with weak, but long–range (LR) correlated disorder, the generic situation in crystals with topological defects like dislocations. We show that the global scenario of the PT differs from that of models with short–range correlated disorder: in addition to the thermal correlation length (which does not diverge, but saturates near T_c) even for very small dislocation densities a second length scale inevitably appears. Its divergence determines the critical point. In the language of the RG, this scale is connected with the existence of a temperature dependent inhomogeneous ground state (GS). This GS is formed by stable nuclei of the ordered phase that appear even *above* the global T_c. We compare our results with experiments for some perovskites and rare earth magnets (REM) [1].

We begin with a brief review of the standard RG treatment of the CB of weakly disordered crystals. The Hamiltonian of the simplest model of this class, for an n–component order parameter φ_i, $i = 1 \dots n$, is given by

$$H = H_{GL}\{\varphi_i\} + \sum_i \int d^3\mathbf{r}\, \frac{1}{2}\, \Delta\tau(\mathbf{r})\varphi_i^2(\mathbf{r}) \qquad (1)$$

with the Ginzburg–Landau Hamiltonian H_{GL} for a pure crystal.

The disorder variable $\Delta\tau(\mathbf{r})$ is usually assumed to be Gaussian distributed, so it is completely characterized by the pair correlator

$$G(\mathbf{r}) = \langle \Delta\tau(\mathbf{0})\Delta\tau(\mathbf{r})\rangle. \qquad (2)$$

This can be described by two parameters, a defect strength $\tau_d^2 = G(\mathbf{0})$ and a single, finite correlation length

R_0, so $G(\mathbf{r}) \to 0$ for $|\mathbf{r}| \gg R_0$. For actual RG calculations a further simplification is usually to ignore the scale R_0 and set

$$G(\mathbf{r}) = u_0\,\delta(\mathbf{r}); \quad u_0 = \tau_d^2\,R_0^d. \tag{3}$$

This is selfconsistently justified with the argument that close to T_c any finite scales like R_0 are irrelevant in comparison to the diverging scale of critical fluctuations, r_c [8].

However, the RG procedure is a description of the critical fluctuations around the GS of a system, and its results are valid only if the GS is determined correctly. Usually, this GS is assumed to be homogeneous; but in fact it has to be determined using either a Landau type saddle point equation for a particular disorder configuration (see below), or the saddle point equation for the disorder averaged free energy. For uncorrelated Gaussian disorder, the latter can be written as

$$(\tau - \Delta)\,\varphi_i^{(0)}(\mathbf{r}) = (u_0 - \lambda_0)\,\left(\varphi_i^{(0)}(\mathbf{r})\right)^3, \tag{4}$$

with the pure vertex λ_0.

If $\lambda_0 \gg u_0$ this equation only has the trivial solution $\varphi_i^{(0)}(\mathbf{r}) \equiv 0$ for $\tau > 0$. But for stronger disorder, $u_0 > u_c = \xi\lambda_0$ with $\xi \approx 1$ nontrivial solutions appear [9], and we have to take into account the presence of a nontrivial GS. The standard RG procedure may be used only if $u_0 \ll \lambda_0$ to describe "weak" disorder in the conventional RG sense [10]. In the opposite case $u_0 > u_c$ a RG calculation has to account for the presence of an inhomogeneous GS.

This leads to the question what kind of GS exists in "high quality" samples of *real* solids. The disorder strength τ_d is small, so a change of the CB can be expected only when the correlation scale R_0 is so large that the approximation of (2) by (3) is no longer valid.

Several types of defects lead to LR correlated random temperature–like disorder. Some special cases, like inhomogeneously distributed point defects have been discussed before [11,12]. But there is a much more generic class of such defects: topological defects like dislocations in crystals. Models with dislocations require a more detailed analysis of many different situations, with varying types of dislocation ensembles and transition scenarios.

We begin our analysis of such crystals with dislocations based on the Landau expansion of the free energy density f,

$$f = H_{GL}\,\{\varphi_i\} + q_{ijkl}\,\epsilon_{kl}^{(d)}(\mathbf{r})\;\varphi_i\varphi_j. \tag{5}$$

The tensor q_{ijkl} describes the coupling [13] between the order parameter and the elastic strain introduced by the quenched disorder, which is given by the sum over contributions from all dislocations [14],

$$\epsilon_{kl}^{(d)}(\mathbf{r}) = \sum_{\alpha,i} \frac{b_\alpha\,s(\phi_i)}{|\mathbf{r}_\perp - \mathbf{r}_i|}, \tag{6}$$

where ϕ_i is the angle between the component of $\mathbf{r}_\perp - \mathbf{r}_i$ perpendicular to the dislocation and the Burgers vector $\mathbf{b}_\alpha$ of a dislocation of type α at $\mathbf{r}_i$, and the sum runs over all dislocations.

To simplify the equations, we will limit the discussion to the case of a scalar order parameter φ and an isotropic elastic medium [13]. The GS for a proper RG treatment can be identified minimizing the free energy (5). This leads to the nonlinear stochastic equation

$$-\,g\;\Delta\varphi + \alpha\;\left(\tau + \frac{q}{\alpha}\;\epsilon_{ii}^{(d)}(\mathbf{r})\right)\;\varphi + \lambda\;\varphi^3 + D\;\varphi^5 = 0. \tag{7}$$

The simplified coupling is $q/\alpha \sim (K/T_0)\;(\partial T_c/\partial p)$ with the elastic modulus K, the Curie temperature T_0 of the undisturbed system and pressure p. The inclusion of a φ^5–term allows us to discuss the weakly first order structural PT in perovskites.

The statistical properties of the solutions of (7) are determined by the random elastic strain $\epsilon = \epsilon_{ii}^{(d)}$. Obviously the coefficient of the linear term in (7) can become negative and allow a nonzero solution of (7), either due to the large contribution to $\epsilon(\mathbf{r})$ of a single dislocation close to $\mathbf{r}$, or because the contributions of several dislocations happen to add up to a large (negative) value of ϵ at $\mathbf{r}$.

For randomly distributed dislocations loops with a radius of curvature L much larger than the average distance r_d between loops [15] the probability distribution function for ϵ in (7) is a Levy distribution [16]: $\mathcal{P}(\epsilon) = \mathcal{P}_{\mu=2}^{(L)}(\epsilon)$, in particular $\mathcal{P}(\epsilon) \sim \epsilon^{-3}$ for $\epsilon \gg 1$. The variance of this distribution is given by

$$\text{var}\;\epsilon \approx (\epsilon(r_d))^2\;\ln(L/r_d) \equiv \left(\frac{\alpha}{q}\right)^2 \tau_1^2 \tag{8}$$

with $\epsilon(r_d) \approx b/2\pi\,r_d$ and the corresponding temperature scale τ_1.

The pair correlation function for this distribution for $r \le L$ is $G(r) = \langle\epsilon(0)\,\epsilon(\mathbf{r})\rangle \sim n_d\,\ln(L/r)$ with the dislocation density $n_d = r_d^{-2}$. It is determined by the contribution of many dislocations on scales $r_{min} < r < L$, while for distances $r < r_{min} = r_d\,(\ln(L/r_d))^{-1/2}$ the random value of $\epsilon(\mathbf{r})$ is dominated by the contribution of the single nearest dislocation.

As the elastic strains are strongest in the immediate vicinity of the dislocations, nonzero solutions of (7) may first appear there as the temperature is lowered from high values. In a first approximation, we can limit (7) to include only the strain induced by the closest dislocation i near $\mathbf{r}$:

$$-\,g\;\Delta\varphi + \alpha\;\left(\tau + \frac{q}{\alpha}\;\frac{b\,s(\phi)}{|\mathbf{r}_\perp - \mathbf{r}_i|}\right)\;\varphi + \lambda\;\varphi^3 + D\;\varphi^5 = 0. \tag{9}$$

The temperature T_n at which nonzero solutions of this nonlinear, deterministic equation first appear can be found rigorously [17] by an analysis of the linear part of this equation. The main results of this analysis are as follows. The reduced temperature $\tau_n = (T_n - T_0)/T_0$ and the characteristic scale of ordered nuclei $R(\tau_n)$ are,

$$\tau_n \approx \frac{1}{\alpha g}\,(q\,b)^2 + \Delta\tau_h, \tag{10}$$

$$R(\tau_n) \approx \frac{g}{q\,b} \approx \sqrt{\frac{g}{\alpha\tau_n}} = r_c(\tau_n). \tag{11}$$

Here $\Delta\tau_h \ll \tau_n$ describes the temperature range of the hysteresis in a crystal without dislocations and a weakly first order PT like the perovskites that display the two length scales [18]. The second expression for $R(\tau_n)$ reflects the fact that nonzero solutions appear when the energy gain from the attractive potential balances the "kinetic" energy due to the gradient term in the free energy.

The above reasoning depends on two assumptions: the possibility to include only the simplest gradient term in (7) and (9), and the validity of the single defect approximation (9).

Obviously, the former requirement holds if $R(\tau_n) \gg a$, where a is the lattice constant. The coefficient α can be written as $\alpha = T_0/C$ with the material's Curie constant C. In perovskites, for displacive PTs [18] $C \approx 10^4$ – $10^5 K$, so $\alpha = O(10^{-2}$ – $10^{-3})$ and, indeed, $R(\tau_n) \approx a/\alpha \gg a$. The scattering experiments for $SrTiO_3$ also show [19] that in the relevant temperature range $r_c \gg a$, so the use of (7) and (9) is justified.

In REM, $C \ll T_0$ [20], so α is large and (7) and (9) cannot be used at all. However, in both cases below T_n the radius $R(\tau)$ grows much larger than r_c, so one can use the estimation

$$R(\tau) \approx \frac{K}{T_0}\,(\partial T_c/\partial p)\,\frac{b}{2\pi\,\tau} \tag{12}$$

in the temperature range where $R(\tau) \gg r_c(\tau)$. Obviously the "divergence" of the scale $R(\tau)$ is described by an exponent $\nu = 1$. The prefactor in (12) has very similar numerical values in perovskites and REM, which leads to scales of a few 10^3 Å in both classes [21]. In the temperature range where (12) is valid, the integrated intensity of scattering from the nuclei of the ordered phase is $I(\tau) \sim n_d R^2(\tau) \sim \tau^{-2}$.

As the temperature is reduced the size and the number of nuclei grow. To determine the global character of the PT one must then investigate the temperature dependence of the disorder averaged GS correlator $g(\mathbf{r}) = \langle \varphi^{(0)}(\mathbf{0})\ \varphi^{(0)}(\mathbf{r})\rangle$ for $r \to \infty$. For this purpose it is crucial to analyze the state of "bridges" with $\tau(\mathbf{r}) < 0$ which link two nuclei that have appeared at some higher temperature [22].

It is easy to show [23,21] that if on such a bridge $\tau(\mathbf{r}) < -\tau_2 \equiv g/\alpha\, r_{min}^2$ the correlation radius of local thermal fluctations is $r_c(\mathbf{r}) \ll r_{min}$. In such bridges the energy of a domain wall (DW) is much larger than T_c, so no DWs will appear. If $-\tau_2 < \tau(\mathbf{r}) < 0$ then $r_c(\mathbf{r}) \geq r_{min}$ and DWs are easy to form. The probability for this is $Prob(r_c(\mathbf{r}) \geq r_{min}) \approx (\tau_2/\tau_1)^{1/2} \equiv z$. The sign of the order parameter in the nuclei connected by a bridge is the same with a probability $1 - z \approx 1$ for $z \ll 1$. Of course, this sign will fluctuate in time as long as overlapping nuclei form finite clusters.

As the temperature is decreased, these clusters will grow, and at T_c an infinite percolating cluster will appear. On this cluster the sign of the order parameter cannot change anymore — this is the spontaneous symmetry breaking that defines the phase transition in the system. Therefore the global PT scenario is a percolative one. The condition $z \ll 1$ can be rewritten as $R(\tau_n) \ll r_{min}$ using (11). This turns out to be equivalent to the condition under which the first nucleation of the ordered phase near dislocations can be described in the single defect approximation (9).

The smaller length scale seen in experiment corresponds to the fluctuations $\tilde{\varphi}(\mathbf{r}) = \varphi(\mathbf{r}) - \varphi^{(0)}(\mathbf{r})$ around the inhomogeneous, temperature dependent GS $\varphi^{(0)}(\mathbf{r})$ determined by (7) [24]. Assuming mean field exponents for simplicity, the width of the corresponding "broad component" in momentum space can be estimated as

$$(\Delta q)^2 = \langle r_c^{-2}\rangle = A_+ \int_0^\infty \tau'\,\mathcal{P}(\epsilon = \tfrac{\alpha}{q}(\tau' - \tau))\,d\tau' + A_- \int_{-\infty}^0 |\tau'|\,\mathcal{P}(\epsilon = \tfrac{\alpha}{q}(\tau' - \tau))\,d\tau' \tag{13}$$

with $\tau = (T - T_0)/T_0$. This implies that the line width remains finite at all temperatures, with a minimal value $\Delta q_{min} \approx r_c^{-1}(\tau_1)$. This is reached (in the mean field approximation, $A_-/A_+ = 2$) at $p = 1/3$. Because in three dimensions the percolation concentration $p_c < 1/3$, this minimum lies slightly below the temperature T_c where the length scale connected with the narrow component of the scattering diverges, as observed in experiments [25].

If $R(\tau_n) > r_{min}$, neither the single defect approximation nor the simple percolative picture of the PT hold, as the inhomogeneous GS $\varphi^{(0)}(\mathbf{r})$ and thermal fluctuations both play an equally important role. In this case a completely new generalized RG scheme is needed.

To summarize, we have analyzed the CB of solids with LR correlated quenched disorder taking into account the appearance of an inhomogeneous, temperature dependent GS. In real crystals the appearance of such a GS is a generic phenomenon due to the almost unevitable presence of dislocations. The appearance of this GS is important even for a qualitative discussion; in particular it leads to the breakdown of the conventional RG procedure.

We have shown that if the condition $R(\tau_n) \ll r_{min}$ holds the global PT scenario consists of two stages: at $\tau = \tau_n$ nuclei of the low-temperature ordered phase appear next to individual dislocation lines. At lower τ

the nuclei grow and begin to overlap; the typical size of magnetically ordered domains is described by percolation theory. It is especially remarkable that this PT scenario becomes more likely when n_d is reduced, because $r_{min} \propto n_d^{-1/2}$. So generally speaking the two length scale phenomenon should be observable in scattering experiments in many crystals. Of course a high resolution setup with sufficient sensibility as used in refs. [3,25,7] is necessary, especially for *better* samples with small n_d, because the volume occupied by nuclei of the ordered phase becomes noticable at temperatures of order $\tau_1 \sim n_d^{1/2}$ (using (8) and (12) one can write $\tau_1 = (n_d\, b^2)^{1/2}$) for most structural PTs and the PT in REM.

The universality of the percolation PT scenario due to LR correlated disorder can be illustrated by the the recent observation [26] of two scales in the neutron scattering data for the Invar alloy $Fe_{1-x}Ni_x$ which is near a morphology boundary for $x \approx 0.3$. Contrary to earlier speculation [8] the description of the CB of *real* crystals very close to T_c must account for LR correlated disorder in addition to the uncorrelated disorder covered by the conventional theory. In fact, the asymptotic CB of "high quality" samples of perovskites and REM is the same as that of a "dirty" alloy with LR chemical disorder [27].

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